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13. ABSTRACT (Maximum 200 words) Single mode cw and pulsed distributed feedback (DFB) QC lasers with wide current tuning and high single mode power, have been realized at $\lambda \approx 4.6 \mu\text{m}$; $5.2 \mu\text{m}$, $8 \mu\text{m}$ and $9.5\text{--}10 \mu\text{m}$ and their trace gas sensing ability (parts per billion in volume or less) has been demonstrated by a variety of spectroscopic techniques. These devices, free-running and stabilized, had cw linewidths of $\sim 1 \text{ MHz}$ and $\sim 10 \text{ KHz}$, respectively. Optical powers in excess of 0.5 W (1 W) at 300 K (200 K) was obtained in 75-stage structures; the cw operating temperature was increased to 175 K using epitaxial-side heat sinking. New chirped superlattice active region designs yielded greater optical power per stage and allowed extension of the operating wavelength to $19 \mu\text{m}$. These longwavelength QCLs use metal semiconductor waveguides supporting a surface plasmon mode, which allows greater optical confinement, and hence lower thresholds than conventional semiconductor waveguides of nearly double thickness. A QCL lasing simultaneously at two or more widely spaced wavelengths and a bidirectional QCL emitting different wavelengths for opposite bias polarity were demonstrated. Gain switching and active modelocking of QC lasers at 5 and $8 \mu\text{m}$ gave pulse widths of 90 ps and a few ps, respectively.					
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Enclosure 1

1. Statement of the problem studied

On August 1, 1998 we entered into a contract (DAAG55-98-C-0050) with DARPA/ARO, entitled Widely Tunable and High Power Mid-infrared Quantum Cascade Lasers. The program ended on July 31, 2000. The goal of this program was to develop widely tunable single mode QC lasers and high power QC lasers operating cw and pulsed with power level in the hundreds of mW to ~ 1 W range at selected wavelengths in the two atmospheric windows (3-5 μm) and (8-13 μm).

2. Summary of the most important results

1) *Widely tunable single mode, narrow linewidth QC-DFB lasers for gas sensing.*

High performance QC-Distributed Feedback (DFB) QC lasers were designed and demonstrated for operation at wavelengths in the two atmospheric windows. This is significant for DOD applications in areas such as detection of weapons of mass destruction in the battlefield and for treaty verification purposes, counter-terrorism and civil defense. Law enforcement and commercial gas sensing application would also benefit. This study targeted three regions near 4.6-4.7 μm ; 7.9-8.5 μm and 9.5-10.5 μm . To achieve high single mode performance, the design for the shorter of these wavelengths utilized a strained AlInAs/GaInAs active region with deep quantum wells (0.7 eV). This made it possible to obtain single mode powers of ~ 100 mW pulsed at room temperature and in cw. This performance was also achieved with an improved top complex grating design, which relies on the periodic modulation of the real and imaginary parts of the mode refractive index as well of the confinement factor. Similar high performance was obtained in the 7.9-8.2 μm range, using in this case a lattice matched active region composition, with a vertical transition 3-well design, as in the shorter wavelength case. Both structures showed significant tuning (~ tens of nm) by ramping linearly the drive current at slow rates. This tuning is caused by heat induced refractive index changes, which changes the wavelength at which the Bragg condition for the grating is satisfied. The ~ 8 μm QC-DFBs were provided to several groups who demonstrated gas sensing with high sensitivity (see later section). In a parallel effort, the wavelength range of the CO₂ laser was targeted. It was shown that the full operating range plus the tuning gaps of this laser could be covered by a mere 10% variation of the QCL active region quantum well thickness. The other fine-tuning is provided by the choice of the position of the Bragg wavelength (i.e the grating period) with respect to the peak of the gain spectrum. The latter work shows that compact QC lasers with power levels of a ~ 1 W or more in cw and pulsed operation are definitely within the reach, thus providing a compact and portable alternative to CO₂ lasers.

Frequency stabilization of mid-infrared quantum cascade (QC) lasers to the kiloHertz was accomplished using electronic servo techniques. Using this active feedback, an 8.5 μm QC DFB laser was locked to the side of a roto-vibrational resonance of nitrous oxide (N₂O). A stabilized frequency noise spectral density of 42 Hz / $\sqrt{\text{Hz}}$ was measured at 100 kHz: the calculated laser linewidth was 12 kHz. For a free running QC laser the linewidth was ~ 1 MHz over a similar time interval of a few ms, limited by the technical noise of the drivers.

2) *High power and improved temperature performance*

The cascading scheme is a characteristic feature of quantum cascade (QC) lasers. It implies that electrons above threshold generate one photon per active region they successively traverse. We have reported a study of the cascading behavior as a function of the number N of stacked active regions. Experimental results were obtained for devices with N = 1, 3, 6, 12, 20, 30, 45, 60, and 75 active stages. The highest optical power and lowest threshold current density was obtained for laser devices with N as high as possible. However, the lowest threshold voltage and the lowest dissipated power at laser threshold were achieved for N = 3 and N = 22, respectively. We further presented the highest power QC lasers so far, which, using N = 75 stages, showed in pulsed mode peak powers of 1.4, 1.1, and 0.54 W at 50 K, 200 K, and room temperature, respectively. Finally, we also demonstrate the first few-stage (N < 10) QC lasers. These QC lasers showed strongly reduced operating voltages. A threshold voltage around 1.5 V was achieved for N = 3. This makes the lasers very well compliant with conventional laser diode drivers, which in turn will simplify their immediate use in systems and applications.

To improve the temperature performance, the epilayer-side mounting of quantum cascade (QC) lasers were demonstrated for better heat sinking. Operated in continuous-wave (CW) mode, these lasers are superior to substrate-bonded devices. The maximum CW temperature is raised by 20 K (up to 175 K) and, at comparable heat sink temperatures, the performance with respect to threshold current, output power, and slope efficiency is greatly improved for the epilayer-side mounted devices. Modeling of the temperature distribution inside the QC laser shows a strong temperature gradient within the active waveguide core, which partly explains the still low maximum CW operating temperatures.

3) *Superlattice QC lasers for long-wavelength operation and higher dynamic range and Surface Plasmon lasers*

In the course of our previous DARPA contract, entitled Developing Thermoelectrically Cooled Continuous Wave Mid-Infrared Quantum Cascade Lasers (DAAH04-96-C-0026), we demonstrated a new structure with a superlattice active region. We recognized that such a design would provide potentially very high current drive capabilities, due to the wide minibands, as well as high oscillator strength, particularly at longer wavelengths. The original structure had doped active superlattices, which hindered low threshold operation. We therefore, in this new contract, concentrated on new designs with *undoped* superlattice active region for narrower gain spectra and hence lower threshold. Two approaches have been demonstrated in the present contract. In the first one a modulation doping scheme is used that creates an electric field, via charge transfer, which effectively cancels the applied field in the active region, thus permitting to avoid field induced localization of the superlattice states. To avoid phonon bottleneck effects, this structure was optimized by making the separation of the first two superlattice states of the second miniband equal to one optical phonon (~ 30 meV). This ensures that the population inversion is concentrated across the high matrix element transition at the miniband edge. In this way an optical power level of ~ 100 mW at ~ 7.9 μm wavelength was achieved with 25-30 stages at 300K. Room temperature operation had not been possible with doped superlattice active region QC lasers.

Even better results, in terms of pulsed threshold at 300 K and optical power, were obtained by designing the undoped active regions as chirped superlattices; i.e. with spatially varying period and duty cycle. In this structure for an applied bias well below threshold, the states of the individual quantum wells comprising the superlattices are not energetically lined up and they remain localized. As voltage is increased near flat band (near the threshold condition), the states line-up and anticross forming bands of states (minibands), which allows high-current injection and laser action with relatively low threshold, due to the reduced broadening of the laser transition. In this way, record peak optical powers of 200 mW at 300 K near 8 μm , with average powers of 15 mW at a few % duty factors, and a threshold of only 5 kA/cm², was demonstrated using only 20 stages.

The chirped superlattice QC laser concept was successfully extended to longer wavelengths: 17 and 19 μm . These are the longest wavelength semiconductor lasers to have ever been demonstrated with III-V semiconductors. This design also made possible for the first time cw operation at 11 μm with power levels of ~ 100 mW at ~ 100 K in QC lasers. Laser action at 19 μm necessitated innovation also in the waveguide design. Essentially, because of the relative small refractive index difference between core and cladding layers, the optical confinement factor is $\Gamma \sim 0.4$ at these wavelengths and requires prohibitively thick epitaxial layers, from a growth point of view. This value of Γ is also hardly sufficient for the modal gain to equate the large optical losses (due to free carrier absorption) at these long wavelengths and achieve threshold. To solve this problem we replaced the conventional dielectric waveguide used in QC lasers at shorter wavelengths (< 17 μm) with a metal semiconductor one, by essentially replacing the upper several microns thick AlInAs cladding layer with a metal film of small skin depth. In this guide, because of the TM polarization of the QC laser radiation, the propagating laser mode is a surface plasmon wave peaked at the metal semiconductor interface that dies rapidly into the semiconductor. In this way, optical confinement factors approaching unity (~ 0.9) were achieved at 19 μm wavelength with only about 4 μm of epitaxial material, as opposed to the more than 8 μm required for a dielectric waveguide. Pulse peak power of tens of mW were achieved, with maximum operating temperatures of ~ 100 K. Single mode operation was demonstrated at 17 μm with such as surface plasmon design, by using as a complex grating an alternation of two metals (Ti and Au) laid on top of the active region, which periodically modulates the skin depth and the refractive index.

4) *High speed operation of QC lasers: gain-switching and active modelocking*

We have reported the first gain switching of QC lasers. In our experiments, a commercial comb generator was driven with approximately half a Watt of rf power from a low phase-noise synthesized signal generator, giving rise to a train of 140-psec wide electrical pulses; the repetition rate is set by the driving frequency (100 MHz). The output of the comb generator was then combined with a dc current in a standard bias 'tee' to drive the laser. Several QC lasers, emitting at ~ 8 and ~ 5 μm (chosen as representatives of the two atmospheric windows), were gain-switched with this approach. In particular, we used two 8 μm lasers based on the 'vertical' transition design with a three-coupled-quantum-well active and a similar 5 μm laser. The laser samples were processed into standard ridge waveguide structures and cleaved into 1.5-mm long and 2.25-mm long bars. The output pulses were measured directly with a fast HgCdTe photovoltaic detector mounted in a liquid-nitrogen dewar, having a specified 3dB cut-off frequency of approximately 1.5 GHz. The pulse width (full-width-at-half-maximum) of the 8 μm lasers was about 200 psec, broadened by the detector response time. The peak power was 12 mW. Shorter pulses (90 ps, deconvolved down to 45 ps, by taking into account the bandwidth of the measuring apparatus) were measured with bound-to-continuum quantum well infrared detectors, provided by H. C. Liu of the National Research Council in Ottawa (Canada).

Active modelocking was demonstrated by driving the cw QC lasers at 5- μm and 8- μm with an RF signal tuned to the separation of the longitudinal modes of the cavity (12.5 GHz). In the absence of optical correlation traces, difficult to obtain in the mid-ir due to the small conversion efficiency of nonlinear crystals, we nevertheless directly inferred modelocking from the abrupt appearance of tens of longitudinal lasing modes of similar heights in the FTIR spectrum, when the driving frequency matched the mode separation and by the high stability of the signal from the QWIP and its ultra-narrow spectrum, as recorded by a spectrum

analyzer. These are the first mid-ir semiconductor lasers to be modelocked. The envelop of the FTIR spectrum showed a series of pronounced maxima as the drive current is increased, which is indicative of self-phase modulation of the pulses. This effect is due to the giant optical Kerr effect, associated with the large nonlinear refractive index of the intersubband transition. From these envelopes we were able to estimate pulse width of 3-5 ps.

5) *Band-structure engineered devices: bi-directional and multiwavelength QC lasers*

A semiconductor laser capable of operating under both positive and negative bias voltage was reported (bidirectional QC laser). Its active region behaves functionally as two different laser materials, emitting different wavelengths, depending on the design, when biased with opposite polarities. This concept has been used for the generation of two wavelengths (6.3 μm and 6.5 μm) in the mid-infrared region of the spectrum from a single quantum cascade laser structure. The two wavelengths are excited independently of each other and separated in time. This may have significant impact on various semiconductor laser applications including trace gas analysis in remote sensing applications using differential absorption spectroscopy.

In another important development, a superlattice active region device, designed to emit simultaneously two widely separated wavelengths, was demonstrated. To accomplish that, the superlattice was engineered so as to have two optical transitions with comparable oscillator strengths, with the upper states separated by less than one optical phonon in which electrons are injected, thus maintaining a similar electron population.

6) *Trace gas-sensing applications.*

Most trace gases of importance in various applications have characteristic vibrational absorption features in the mid-infrared, between 3 and 17 μm wavelength. As discussed before, QC-DFB lasers can be designed to emit at any wavelength in this spectral range. Therefore, it was straightforward to examine the lasers' potential in demanding gas-sensing applications in collaboration with many groups in academia, industry and national labs. Overall these results reported below illustrate the potential of QCLs in gas sensing with sensitivities down to a few parts-per-billion-in-volume (ppbv) or better, depending on the technique used. Particularly attractive is the use of room temperature/thermoelectrically cooled pulsed QCLs for portable and compact sensors.

D. Sonnenfroh and coworkers of Physical Sciences Inc. have used low duty cycle ($< 1\%$) pulsed room temperature 5.4 μm QC-DFB lasers and balanced ratiometric detection in a direct absorption experiment of trace gases. They demonstrated sensitivities for N_2O of 10 ppm-m and for NO of 55 ppb-m. These results are very promising for the realization of sensitive, very compact and portable sensors for real-world applications.

S. W. Sharpe et al. at the Pacific Northwest National Laboratory (PNNL), WA, conducted high-resolution, Doppler-limited, direct absorption measurements of NO and NH_3 using QC-DFB lasers at 5.2 and 8.5 μm , respectively. The laser drive current was a saw-tooth ratchet with 6 – 11 kHz repetition rate; with the rising current the laser would tune $\sim 2.5\text{ cm}^{-1}$, covering e.g. 11 absorption features of NH_3 . The noise equivalent sensitivity limit in these measurements was 3×10^{-6} absorbance.

The group of Dick Zare at Stanford University, CA, together with J. Oomens et al. at the University of Nijmegen, The Netherlands, reported photoacoustic spectroscopy on NH_3 and H_2O diluted in N_2 using a cw QC-DFB laser emitting at 8.5 μm wavelength. The noise-limited minimum detectable concentration of NH_3 was 100 ppbv for 1 s integration time. B. Paldus and coworkers at Informed Diagnostics, along with Dick Zare, have also demonstrated sub-ppbv (NH_3) sensitivity measurements using cavity ring down spectroscopy.

The groups of Bob Curl (Nobel Laureate in Chemistry) and Frank Tittel at Rice University, TX, reported on measurements of the concentration of $^{12}\text{CH}_4$, its natural isotopes $^{13}\text{CH}_4$ and $^{12}\text{CH}_3\text{D}$, H_2O , N_2O , and $\text{C}_2\text{H}_5\text{OH}$ diluted in standard air using a direct absorption technique around 7.95 μm wavelength. With a 100m multi-pass gas cell and employing a new background subtraction method, they were able to demonstrate a sensitivity limit in the ppbv concentration range, e.g. 125 ppbv of $\text{C}_2\text{H}_5\text{OH}$.

Bill Weber and coworkers at the Ford Research Laboratories have used a quantum cascade distributed feedback laser operating at 5.2 μm to obtain sub-Doppler resolution limited saturation features in a Lamb-dip experiment on the $\text{R}(13.5)_{1/2}$ and $\text{R}(13.5)_{3/2}$ transitions of NO. The laser is operated CW in a LN_2 Dewar. Lamb dips appear as transmission spikes with full widths of $\sim 4.3\text{ MHz}$. At this resolution, the 73 MHz lambda doubling of the $\text{R}(13.5)_{3/2}$ line, which is normally obscured by the 130 MHz Doppler broadening, is easily resolved.

C. Webster and coworkers at JPL, California Institute of Technology, conducted measurements of the concentration of CH_4 and N_2O in the earth's atmosphere from ground level to the stratosphere ($\sim 70,000\text{ ft}$) using a cw operated 7.95 μm QC-laser and a wavelength modulation technique. The laser in a LN_2 dewar was on board a high-altitude air-plane; the surrounding air would be sucked into a multi-pass gas cell aboard the plane, as the plane makes 8 hr long flights to map the atmosphere for the trace gases. The noise-equivalent sensitivity limit was $\sim 2\text{ ppbv}$.

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4. List of participating scientific personnel

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5. Inventions

- a. Chirped superlattice active region QC lasers
- b. Multiwavelength QC lasers
- c. Bidirectional QC lasers
- d. QC laser with electric field-free superlattice regions
- e. Surface plasmon QC lasers

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